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VOLATILITY OF AEROSOLS IN THE WESTERN EUROPEAN ENVIRONMENT

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FINAL TECHNICAL REPORT

S.G. Jennings

December 1989

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60 D D EUROPEAN RESEARCH OFFICE

United States Army Research, Development
and Standardization Groups-U.K.
223-231 Old Marylebone Road
London NW1 5TH
England

Contract Number DAJA 45-86-C-0051

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The views, opinions and findings contained in this report
are those of the author and should not be considered as
an official Department of the Army position, policy, or
decision unless so designated by other documentation.

ABSTRACT

Volatile properties of ^{marine} and modified maritime aerosol were measured at the remote site of Mace Head ($53^{\circ}19'N$, $9^{\circ}54'W$) on the west coast of Ireland, located on the eastern edge of the North Atlantic Ocean. The volatility measurements were made with a light scattering counter equipped with a temperature-controlled heated intake. The work extends the temperature range from $300^{\circ}C$ to $850^{\circ}C$ for the first time for aerosol volatility studies. Representative measurements made over the period of nearly two years show that the submicrometer particles with radius < 0.2 micrometer are highly volatile for the maritime aerosol and show temperature-fractionation features of ammonium sulphate (or ammonium bisulphate). It is estimated that 85 - 95 % of this size fraction (by volume) is composed of these sulphates. For the higher temperatures, temperature-fractionation characteristics of sodium chloride are shown for the supermicron and also for the submicron maritime particles. About 80% of the particle size interval of $0.3 - 1.5 \mu m$ radius is composed of sodium chloride. The temperature profile curves for the modified maritime aerosol which has made a partial traverse overland do not display definitive features characteristic of known atmospheric constituents such as ammonium sulphate or sodium chloride but rather properties indicative of mixtures of these species with other unknown constituents. For both air masses, between about 5 to 30 % (by volume) of the aerosol particles remain involatile at least up to a temperature of $850^{\circ}C$.

→ **Keywords:** Temperature, Particle Size, Mixtures, Volatility of Aerosols,
→ Western Europe, Physical Chemistry

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Fig. 6 Aerosol size distributions at Mace Head as a function of temperature up to 400 $^{\circ}\text{C}$.
Measurement Periods: (A): June 1 1988; (B): Sept 20 1988

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Measurement periods: (A): June 1 1988; (B): Sept 20 1988

Fig. 8 Log normal size distribution fits to the ambient (upper curves) and residual (lower curves) aerosol for June 1-2, 1988. The log normal fitted parameters are given in Table 3.

TABLE CAPTIONS

Table 1 : Field measurement data periods.

Table 2 : Mesured particle concentration and volume fraction of volatile aerosol

Table 3 : Log-normal size distribution parameters for the ambient (unheated) and residual (heated), for measurement period June 1-2, 1988.

STATEMENT OF RESEARCH PROBLEM

Until this contractual work was performed, it was only possible to perform aerosol volatility work up to a temperature of 300°C . Therefore the nature of the volatility of atmospheric aerosol particle constituents was unknown beyond this temperature. The research problem associated with this contractual work consisted (a) firstly of testing and designing volatility apparatus in order for it to operate at significantly higher temperatures (of order 850°C) than hitherto possible; (b) secondly of making successful laboratory and field measurements of aerosol volatility at these relatively high temperatures.

SUMMARY OF MOST IMPORTANT RESULTS

Research conducted during the term of the contract included the successful testing of volatility apparatus and extension of its operating temperature up to 850°C (from 300°C). This allowed laboratory temperature fractionation curves to be obtained for an additional range of salts such as sodium chloride, sodium nitrate, sodium carbonate and sodium sulphate.

Aerosol volatility work at Mace Head field research station was carried out for the first time for temperatures up to 850°C. The use of the much higher temperature volatility apparatus allowed the inference and quantification of sodium chloride as a dominant component of the supermicron fraction of both the maritime and modified maritime air masses for the western European environment. In addition the work also showed that sodium chloride is also present in the submicron fraction of the aerosol. For both air masses a sizeable volume fraction (from 5-30%) of the aerosol particles remain nonvolatile-at least up to 850°C.

LIST OF PUBLICATIONS AND PRESENTATIONS

Publications

Jennings S. G. and C.D.O'Dowd, Volatility of aerosol at Mace Head, on the west coast of Ireland, submitted to J. Geophys. Res.

Published Proceedings

Jennings S. G., T.C.O'Connor and C.D.O'Dowd, Volatility of maritime aerosol, in Atmospheric Aerosols and Nucleation, Ed. P.E. Wagner, G. Vali, pp. 257-260, Berlin, Heidelberg, Springer-Verlag, 1989.

Presentations at Scientific Meetings

Jennings, S. G., R. G. Pinnick, and G. Fernandez, Aerosol particle volatility, Aerosol Society Annual Conference, Loughborough, England, April 1987.

Jennings, S. G., and C. D. O'Dowd, Volatility of background aerosol, International Conference on Aerosols and Background Pollution, Galway, Ireland, June 1989.

Holt, E. H., R. G. Pinnick and S. G. Jennings, Comparison of aerosol characteristics in the arid southwestern U.S. and the west coast of western Europe, International Conference on Aerosols and Background Pollution, Galway, Ireland, June 1989.

PARTICIPATING SCIENTIFIC PERSONNEL

Principal Investigator: Dr. S. G. Jennings

Research Scientist: C. D. O'Dowd, B.Sc.

Research Assistant: F. M. Mc Govern, B.Sc., M.Sc.

Research Student: J. Desbonnet

VOLATILITY OF AEROSOLS IN THE WESTERN EUROPEAN ENVIRONMENT

1. INTRODUCTION: The presence of aerosol particles can severely affect the propagation or transmission of electromagnetic radiation in the atmosphere. Quantitative estimates of these effects generally require knowledge of the size distribution, number concentration, complex refractive index and species identification of the aerosol particles. It is well known that the turbid atmosphere containing wet or dry particulate matter adversely affects the performance of electro-optical (E-O) sensors or systems utilized by the U.S. Army. As such, one would like to accurately predict the transmitted and backscattered electro-magnetic energy and other scattering parameters in the presence of aerosol particles. It would be of considerable interest to reliably predict atmospheric propagation from measurable physical parameters of the aerosol.

There is a severe lack of reliable data bases on aerosol properties and constituents particularly in Europe. Recent advances in measurement techniques (Pinnick et al. (1987)) have permitted the determination of the degree of volatility of the aerosol constituents, which allows aerosol species identification to be made.

The work under contract number: DAJA 45-86-C-0051, describes aerosol volatility work carried out primarily at Mace Head, on the west coast of Ireland. Some measurements were also made aboard a research vessel (Friedrich Heincke) out at sea, and approximately due west of the Mace Head site. The work extends the temperature range from 300°C to 850°C for the first time for aerosol volatility studies. In addition, it represents the first volatility work for western European aerosol on traversing the eastern Atlantic ocean. Extensive volatility measurements have already been presented in previous

interim reports, particularly in the 4th., 5th. and 6th. interim reports. This final report will focus on the contrast between volatility measurements for a typical maritime air mass (for the period June 1st.-2nd. 1988) and that for a modified maritime air mass (for the period September 20th. 1988), caused by partial traverse of the original maritime air mass over land.

2. LABORATORY VOLATILITY MEASUREMENTS

2.1 Volatility Instrumentation and Techniques

Details of the volatility instrumentation are described fully in the 5th. interim report. The basic instrumentation consists of a commercial (Particle Measuring System's) light scattering counter, model ASASP-X, to which is attached a temperature-controlled heated inlet. The aerosol particles are drawn through a fused -quartz tube which is heated by means of a nichrome ribbon wrapped around the tube.

2.2 Laboratory Volatility Measurements

Response measurements were made of the heated intake apparatus to polydispersions of previously identifiable atmospheric constituents: ammonium sulphate, ammonium chloride, ammonium nitrate, sodium chloride, sodium carbonate, sodium sulphate, calcium chloride and calcium sulphate. Examples of temperature fractionation curves for some of these laboratory aerosols are shown in Fig. 1. These fractionation curves are used as a data base for comparison with actual field fractionation curves of the natural atmospheric aerosol in order to infer the presence of particular aerosol constituents.

3. VOLATILITY FIELD MEASUREMENTS OF ATMOSPHERIC AEROSOL

The measurements were made at the remote site at Mace Head ($53^{\circ}19'N$, $9^{\circ}54'W$) on the western coast of Ireland, located on the eastern edge of the North Atlantic ocean. The nearest major man-made aerosol sources lie some five thousand km west, along the eastern coast of the United States and Canada. The prevailing (for about 70% of the time) wind flow across Ireland is south-westerly to westerly in direction from the Atlantic Ocean. The site is exposed to about a 120° clean air sector (between about 180° to 300° from N). The research station site is essentially unencroached by land mass, being positioned about four hundred metres from the ocean. There is no human habitation along the prevailing wind direction between the site and the Atlantic ocean and is thus ideally located to measure the natural background aerosol. The nearest metropolis is Galway City, located some 88 km due east of the site. The aerosol sampling took place in proximity to the research station cottage at an elevation of 20m above sea level. The aerosol particle inlet is about 2.5 m above ground.

To ensure that the headed quartz tube at high temperature did not produce extraneous new particles, tests were conducted with particle-free filtered air passing through the heated inlet. The tests proved negative.

The particle sizing instrumentation was placed outside - under ambient conditions-so that the aerosol was sampled as close as possible (to minimise aerosol particle loss) to the inlet of the particle counter. The particle laden air enters a quartz tube of length 45 cm, of which the central portion of 30 cm length is wrapped with a nichrome ribbon. The length of tubing between the quartz tubing exit end and the entrance to the particle counter (see Fig. 1) is minimised to about 16 cm, thus giving a total of 61 cm for total traverse distance of the particles before being sampled by the counter.

Estimates of particle loss in the tubing did not exceed 13% for the largest sized particles, using fractional loss formulae due to gravitational deposition in circular tubes (Davies, 1966).

A summary of the major field data measurement runs made during the contractual period is given in Table 1. Analysis of field data has been already presented in the 4th., 5th. and 6th. interim reports. This final report will focus on a comparison between the volatility characteristics of a maritime air mass with that of a modified maritime air mass. All field volatility measurements were made at Mace Head research station with the exception of the data taken on April 5th.-7th. 1989, aboard the research vessel, Friedrich Heincke.

Two periods are chosen as representative of volatility measurements made over a period from December 1987 to September 1988. The period June 1 -2nd, 1988 is fairly typical of a maritime airmass, as indicated by the air-mass trajectory shown in Fig. 2(A). The period September 20th, 1988 represents a modified maritime airmass as indicated by partial traverse of the air mass overland for the three upper pressure levels, seen in Fig. 2 (B). The trajectories were provided by Joyce Harris of the Geophysical Monitoring for Climatic Change (GMCC) Group at the National Oceanic and Atmospheric Administration (NOAA) at Boulder, Colorado. The trajectory programme used is called "GAMBIT" : Gridded Atmospheric Multilevel Backward Isobaric Trajectories and is described by Harris (1982).

An example of aerosol volatility data taken at Mace Head for the period June 2 1988 is shown in Fig. 3. Plotted are measurements of aerosol temperatures and aerosol particle concentration in four particle size channels versus local time. The measurements represent averages over fairly short time periods (the particle concentration was averaged over 20 degree temperature bins and then shifted by 10 degree increments to the next data bin) causing some statistical

Table 1Field Measurement Data Periods

<u>Date</u>	<u>Prevailing Wind Direction</u>	<u>Comments</u>
Dec. 10-11, 1987	Easterly	Temperature at 300°C solenoid valves in operation. 2 s data throughout
Dec. 16-17, 1987	Southerly	Some measurements bypassing solenoid valves were made
Jan. 13-15, 1988	South-westerly-westerly	Temperature at both 300° and 400°C
March 10-12, 1988	Westerly	4 s data, temperature at 400°C; no solenoid valves in use. Temperature probe on outside of heater tube.
April 19-22, 1988	Southeasterly to southwesterly	No solenoid valves in use, all 400 °C data.
June 1-3, 1988	Southwesterly	900°C data
August 30-31, 1988	Southwesterly	850°C data
Sept. 20-21, 1988	West, Southwesterly and Southerly	850°C data
March 15-16, 1989	Southwesterly	850°C data
April 5-7, 1989	Easterly	850°C data

fluctuations for the largest particle size channel. The modulation of particle concentration induced by heating the aerosol over 60 minute cycles is evident. Reduction in the coarse particle concentration (range 0) to one particle per cm^3 (one count per 4 s interval) is seen for four of the time periods in Fig 3. The reduction in particle concentration down to 10^{-2} for two other time periods on the ordinate of Fig. 3 is a register of zero particle count due to complete volatilization of all the particles for that coarse particle size range. The data is replotted in raw format in Fig. 4 showing the temperature dependence of aerosol concentration. It is seen that the individual curves have approximately the same form which implies that the change in concentration is caused by temperature change and not by natural variations in the aerosol concentration over the 60 minute cycle period.

The data is replotted in Fig. 5(A) where the field data has been averaged over the entire nine cycles (9 hour period) so that only a single fractionation curve up to a temperature of 800 $^{\circ}\text{C}$ results for each particle size range. The resulting average temperature fractionation curves up to about 260 $^{\circ}\text{C}$ for submicrometre particles compare favourably with the laboratory data for ammonium sulphate (and ammonium bisulphate) suggesting these materials are a major constituent of the submicrometre aerosol fraction. A laboratory curve for sodium chloride is also shown. Initial reduction in particle concentration for the largest size range 0 occurs approximately at the same temperature for the field and laboratory data suggesting that sodium chloride is a constituent of the largest size mode. It is also clear that the majority of the submicrometre aerosol is largely unaffected at these high temperatures.

The corresponding plot for the modified maritime air mass is shown in Fig. 5 (B). The shapes of the temperature fractionation curves are quite different from those in Fig. 5 (A). The particles in the smallest size range remain almost involatile up to a temperature of 240 $^{\circ}\text{C}$. There is a gradual fall off

in particle concentration for the next two size ranges beyond the characteristic knee of the ammonium sulphate profile by about 20 degrees indicating that the aerosol is not pure ammonium sulphate. In contrast to the maritime behaviour the particles show a gradual decrease in concentration for all particle ranges with increasing temperature.

Measurements of particle size distribution at Mace Head for both ambient (unheated aerosol) and the heated aerosol are shown in Figs. 6 and 7 for the two different measurement periods referred to earlier. Differential particle number concentration (particles $\text{cm}^{-3} \mu\text{m}^{-1}$) as a function of particle radius (μm) are presented in Fig. 6 for four temperature bins of 10-80 $^{\circ}\text{C}$, 80-180 $^{\circ}\text{C}$, 180-240 $^{\circ}\text{C}$ and 240-400 $^{\circ}\text{C}$ for ambient to 400 $^{\circ}\text{C}$ data for the two measurement periods. The plots represent averages over several cycles, which reduces statistical errors due to characteristic low numbers of particles counts in the larger size categories. For the maritime aerosol the decrease in particle concentration due to particle evaporation or decomposition for the two highest temperature bands is manifest, that is for temperatures $> 180 ^{\circ}\text{C}$. The results generally show the presence of volatile particles for particle radii $< 0.2 \mu\text{m}$ and the larger sized mode of particles (radius $> 0.2 \mu\text{m}$) possessing non-volatile particles in general. Particle volatility extends over a larger particle size range for the modified maritime aerosol, although particle concentration is only significantly decreased for the largest temperature band.

Fig. 7 shows differential particle size distribution data for the ambient to 850 $^{\circ}\text{C}$ temperature range, for four temperature bins of 10 - 100 $^{\circ}\text{C}$, 100 - 180 $^{\circ}\text{C}$, 180 - 700 $^{\circ}\text{C}$ and 700 - 850 $^{\circ}\text{C}$. For both the maritime air mass (Fig. 7 (A)) and the modified maritime air mass (Fig. 7 (B)) the effect of temperature up to 180 $^{\circ}\text{C}$ on particle concentration is negligible. Apart from particle radius $\leq 0.1 \mu\text{m}$ the particles for the modified maritime air mass are more

volatile at the higher temperature bands. The particle concentration for the maritime aerosol, decreases for the highest temperature band suggesting the presence of sodium chloride as also corroborated by Fig. 5 (A).

Quantitative estimates obtained from the differential particle number distribution of number concentration of volatile particles due to heating up to 400 °C and 850 °C for the four size ranges of the particle counter is given in Table 2. The corresponding percentage reduction in aerosol volume during heating is also given in Table 2.

Table 2 Measured particle concentration and volume fraction of volatile aerosol.

(a) Percentage change in aerosol particle number concentration due to heating

Ambient - 400 °C

Ambient - 850 °C

Light scattering counter measurements

Light scattering counter measurements

(Particle radius in micrometres)

(Particle radius in micrometres)

<u>Measurement</u>	0.045-	0.075-	0.12-	0.3-	0.045-	0.075-	0.12-	0.3-
<u>Period</u>	0.1	0.15	0.42	1.5	0.1	0.15	0.42	1.5
June 1.'88	93.8	90.8	49.5	3.9	93.8+0.4	90.9+0.4	66.9+1.3	80.8+2.1
Sep.20.'88	33.8	74.6	88.7	41.4	78.5	94.1	97.4	78.0

(b) Percentage change in aerosol volume due to heating

Ambient - 400 °C

Ambient - 850 °C

Light scattering counter measurements

Light scattering counter measurement

(Particle radius in micrometres)

(Particle radius in micrometres)

<u>Measurement</u>	0.045-	0.075-	0.12-	0.3-	0.045-	0.075-	0.12-	0.3-
<u>Period</u>	0.1	0.15	0.42	1.5	0.1	0.15	0.42	1.5
June 1.'88	94.2	87.1	19.0	1.83	94.0	88.1	67.9	83.5
Sep 20.'88	46.9	80.6	85.2	11.4	84.7	95.7	95.0	75.4

*Typical error bounds are shown for the data of June 1, 1988 for ambient - 850 °C temperature range.

The ambient particle size distribution and in certain cases the residual (after heating) particle size distribution was fitted with log-normal distribution parameters. An example of this procedure is shown in Fig. 8 where the log-normal modes is fitted to the ambient and residual particles (after being heated to 850 °C) for the measurement period of June 1-2, 1988. The values of the log-normal parameters are shown in Table 3. The bumps in the centre of the distribution are probably partially due to the artifact of the ASASP-X instrument which is difficult to accommodate (Garvey & Pinnick, 1983) without knowledge of the aerosol refractive index.

Table 3. Log-normal size distribution parameters for the ambient (unheated) and residual (heated) aerosols, for measurement period June 1-2, 1988.

<u>Fine particle mode</u>			<u>Course particle mode</u>			Integrated ⁺
N (cm ⁻³)	r _g (μm)	σ _g	N (cm ⁻³)	r _g (μm)	σ _g	total mass (μg m ⁻³)
324	0.076	1.35	19.5	0.28	2.08	30.0
30	0.083	1.67	3.4	0.32	1.78	5.1

+ mass calculated from the measured data points, assuming a particle density of 2g cm⁻³ for size range 0 and a particle density of 1.7g cm⁻³ for the remaining 3 size ranges.

4. DISCUSSION

In general, for the volatility data to be meaningful, changes in aerosol concentration over the 60-minute cycle must be small compared to changes induced by heating. Variations in aerosol concentration over these 60-minute temperature cycles for the measurements reported here are estimated to be less than $\pm 20\%$ (see Fig. 3 for example). It is assumed that particle loss is due to evaporation of volatile particles subject to the relatively intense heating. The diffusion of evaporated molecules is such that any gases driven off the aerosol particles during heating recondense on the larger surface area of its unheated tube section rather than back onto the much smaller surface area of the aerosol particles. Estimates of particle loss in the cooling section for laminar tube flow due to thermophoretic forces have been made by Stratmann & Fissan (1988). They find particle penetration of over 80% for temperature differences of 200 °C between the aerosol inlet and the tube wall temperature for particle radii up to 0.035 micrometre. Moreover, the thermophoretic velocity and hence the thermophoretic force is considerably reduced for increasing particle size resulting in increased particle penetration (by an order of magnitude as the particle radius is increased by a factor of 40 from 0.035 μm) as shown by Waldmann and Schmitt (1966). Thus it is considered that particle loss due to thermophoretic forces can be taken as negligible for the range of particle sizes encountered in this work.

Average wind speeds of about 6.5 ms^{-1} prevailed for the two measurement case studies presented here. Since these wind speeds are above the threshold (about 4.0 ms^{-1} (Monahan & O'Muircheartaigh, 1986) for the onset of whitecapping, some unquantifiable amount of aerosol production from the production of jet droplets is likely. Surf zone production of particles is considered to be minimal since surf zone particles tend to consist more of the larger coarse size fraction beyond the sensing range of the particle counter

used here. In addition, the inland shore distance over which surf can occur at the site is relatively short (few tens of metres) and furthermore the aerosol sampling took place, a considerable distance from the surface zone region.

Maritime aerosol sampled on the west coast of Ireland with long trajectory path over the North Atlantic Ocean possesses two main components: a component with particle radius $< 0.2 \mu\text{m}$ consists mainly of ammonium sulphate; and a component with particle radius $> 0.2 \mu\text{m}$ consisting largely of sodium chloride. Below heating temperatures of 400°C there is little variation in particle characteristics for particle radius $> 0.2 \mu\text{m}$. At higher temperatures up to 850°C the greatest reduction in particle number concentration occurs for the largest size range category ($0.3 - 1.5 \mu\text{m}$ radius), reflecting the presence of sodium chloride particles. However some reduction also occurs for the submicrometre fraction (below about $0.2 \mu\text{m}$), indicating the presence also of sodium chloride particles for that lower particle size range. Between about 10 - 33% of the particles remain as involatile particles after heating to 850°C , which can be attributable to residue quartz, calcite and clay mineral particles, as evidenced by supplementary X-ray dispersive analysis of single submicron and supermicron particles collected onto nuclepore filters (presented in the 6th Interim Report).

The modified maritime aerosol with a trajectory partially overland shows different volatility characteristics as compared to the pure maritime aerosol. At lower temperatures ($< 400^\circ\text{C}$) a much smaller percentage of the modified aerosol particles are volatile for the smallest size range as seen in Fig. 6 and Table 2. This could be attributable to the presence of carbon particles. For larger sizes the degree of volatility is greater for the modified maritime particles, which as yet remains unexplained. Particle volatility is about the same for both air masses for the larger size range which probably reflects the

dominant presence of sodium chloride particles in both cases. Between about 10 to 20 percent of number concentration of non-volatile particles (non-volatile at least up to 850 °C) exists for both of the air masses investigated. Further work is required to identify the nature of both volatile and non-volatile particles in order to differentiate more fully between aerosol particles of different air mass types and air mass histories.

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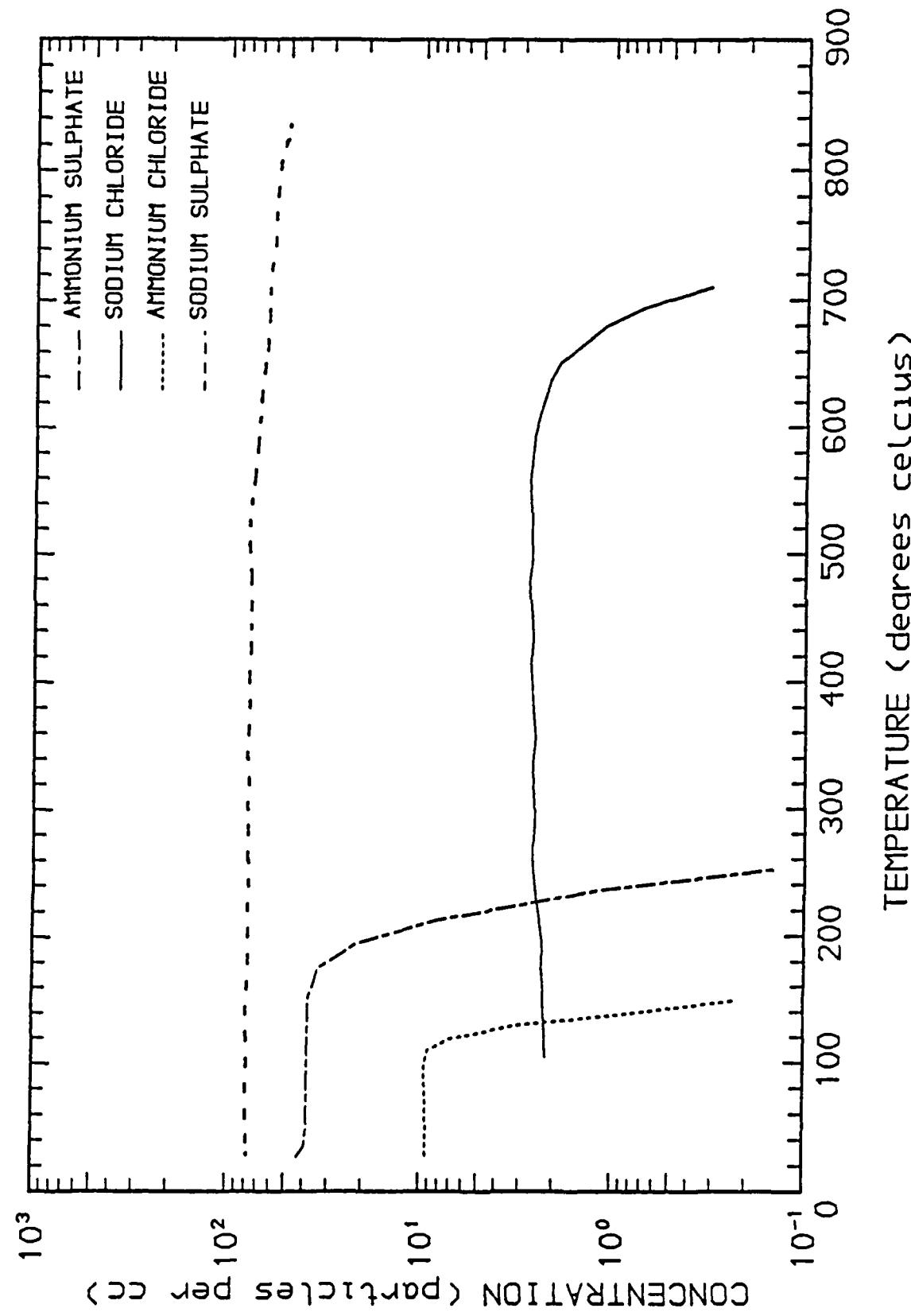


Fig. 1 Laboratory aerosol temperature fractionation curves for ammonium sulphate, ammonium chloride, sodium chloride and sodium sulphate.

TRAJECTORIES TO MCH (53.32N, 9.85W)

88153 - 6/ 1/88 AT 12Z

A: 1000 MB B: 850 MB C: 700 MB D: 500 MB

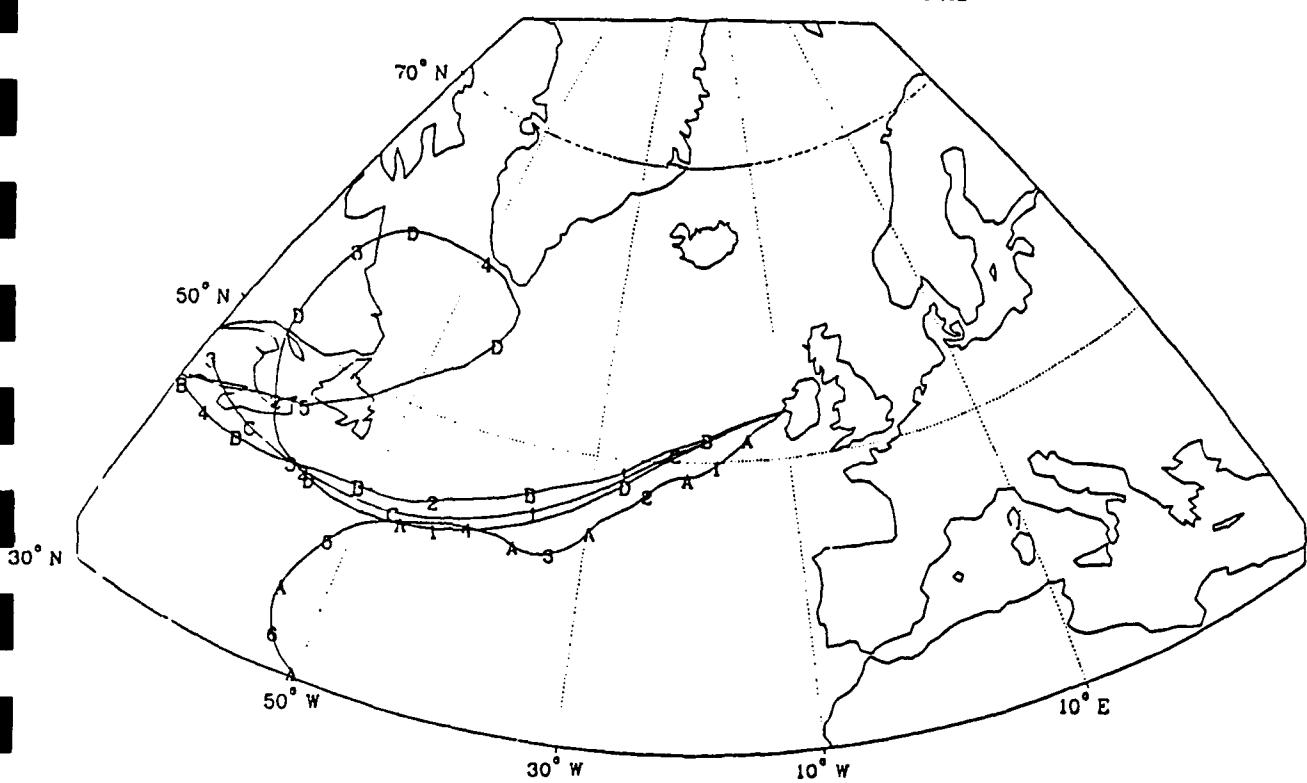
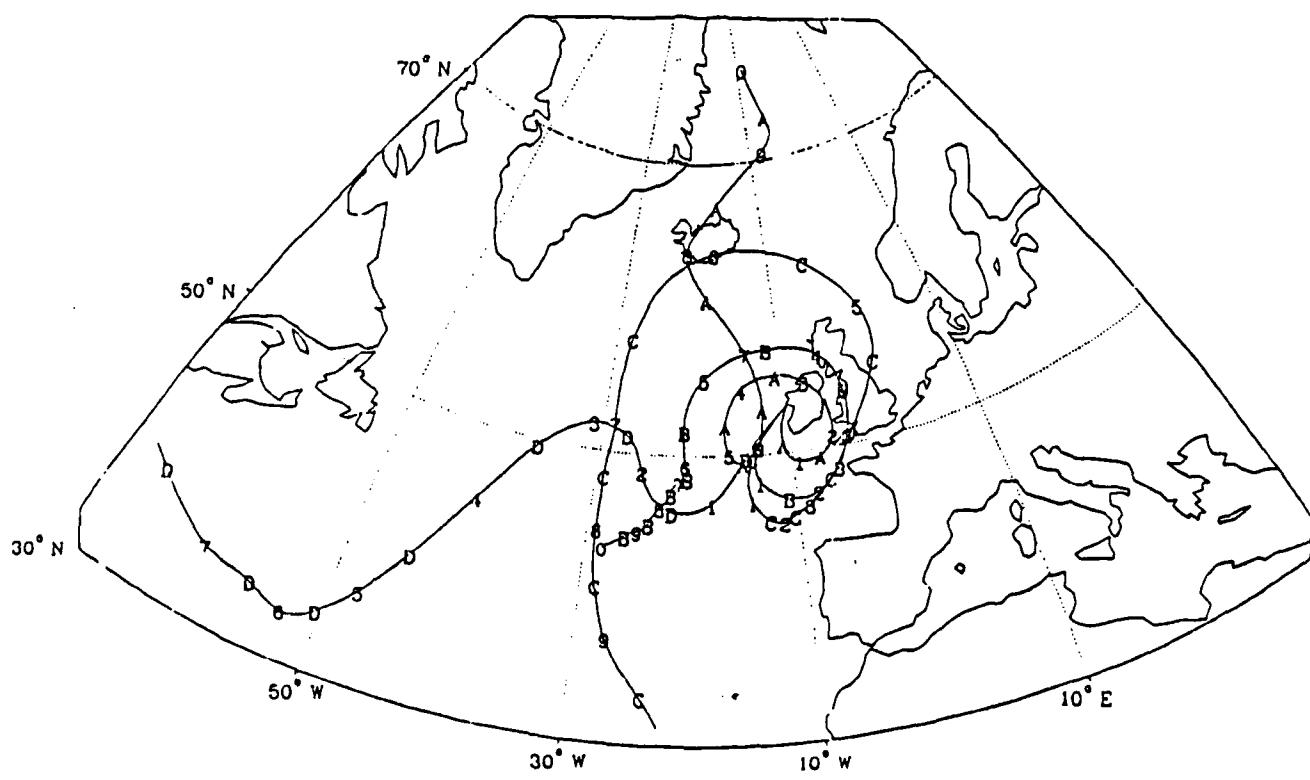


Fig. 2 Atmospheric air trajectories to Mace Head (MCH) on June 1 1988 and on September 20 1988, for pressure levels of 1000 mb, 850 mb, 700 mb and 500 mb. The numbers on each trajectory represents the number of days the air mass is from Mace Head.

TRAJECTORIES TO MCH (53.32N, 9.85W)

88264 - 9/20/88 AT 0Z

A: 1000 MB B: 850 MB C: 700 MB D: 500 MB



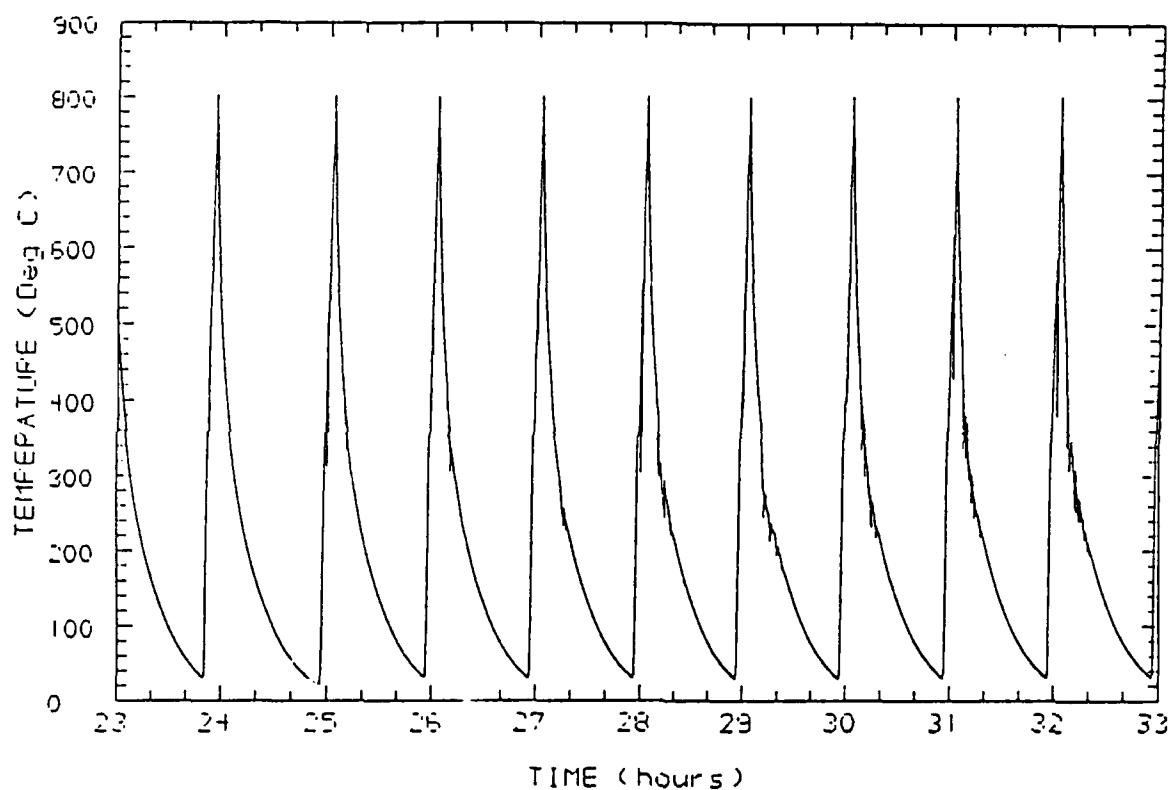
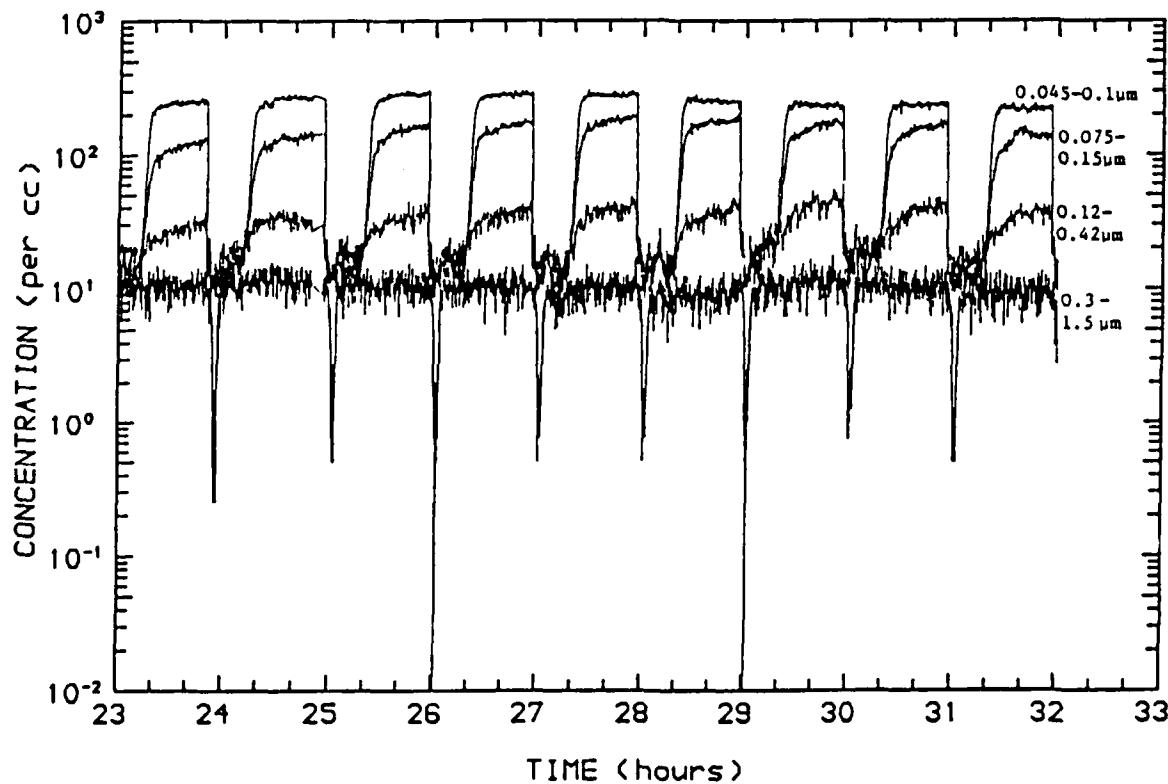


Fig. 3. Aerosol volatility measurements made at Mace Head field site. Temperature and aerosol concentration per cm^3 in four particle size ranges (given for particle radii 1 μm) is plotted versus local time for the period June 2 1988.



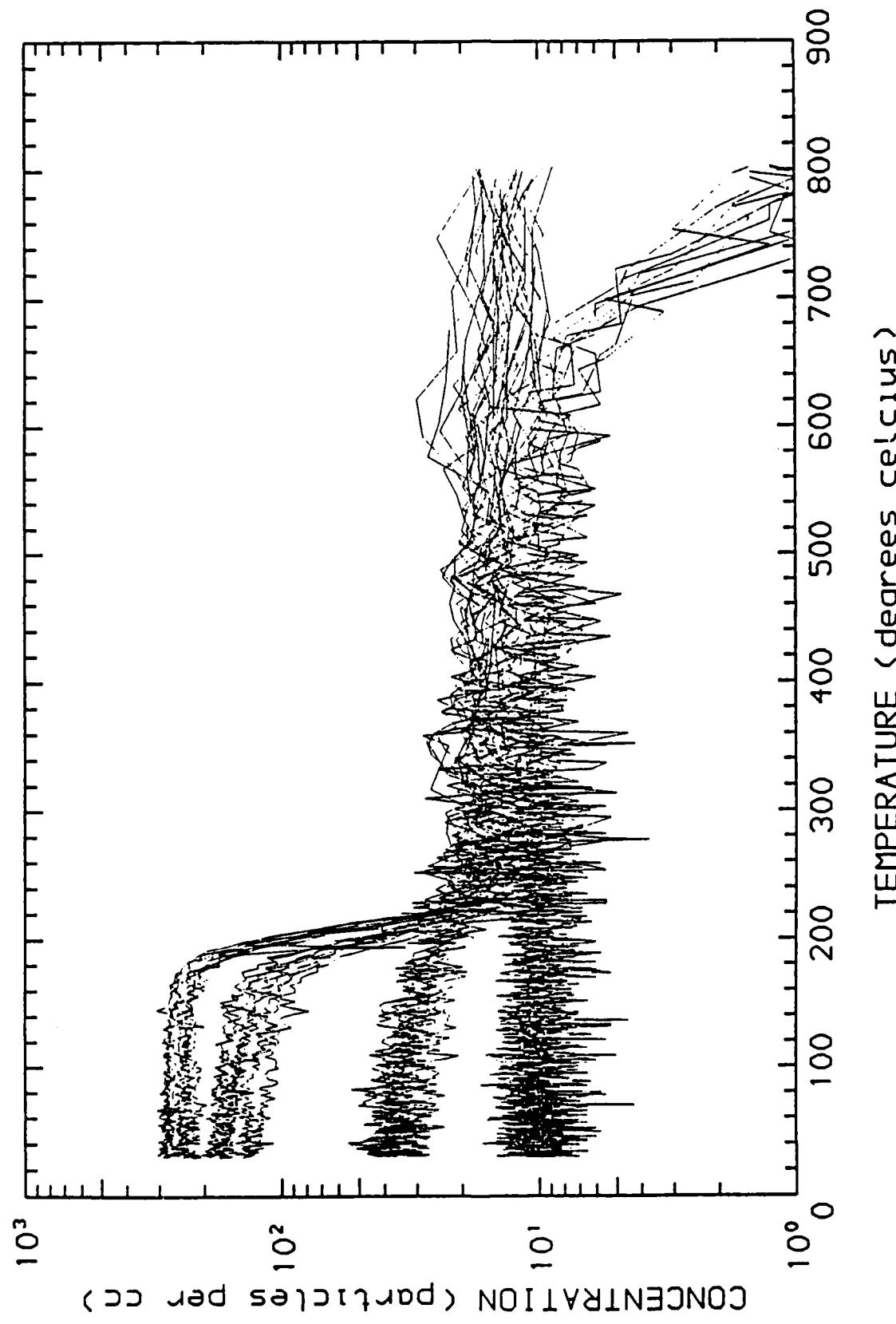


Fig. 4. Volatility measurements of Fig. 3 replotted in raw data format as temperature fractionation curves.

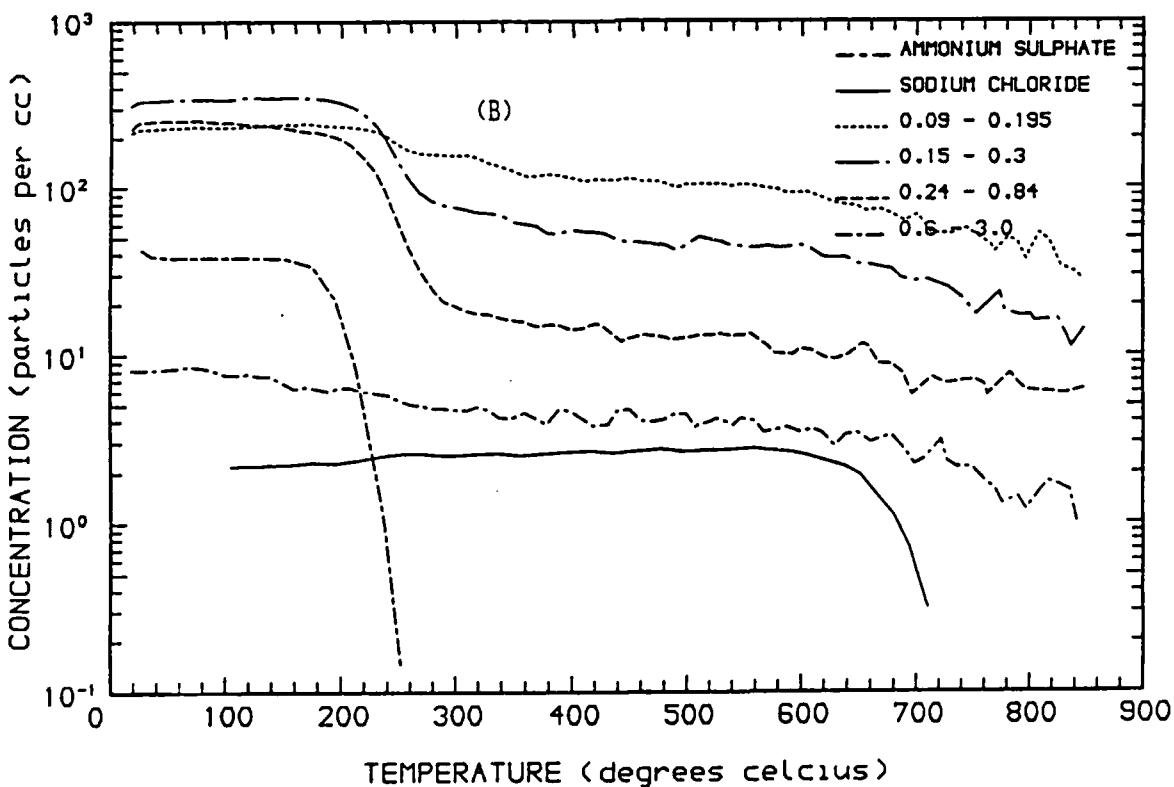
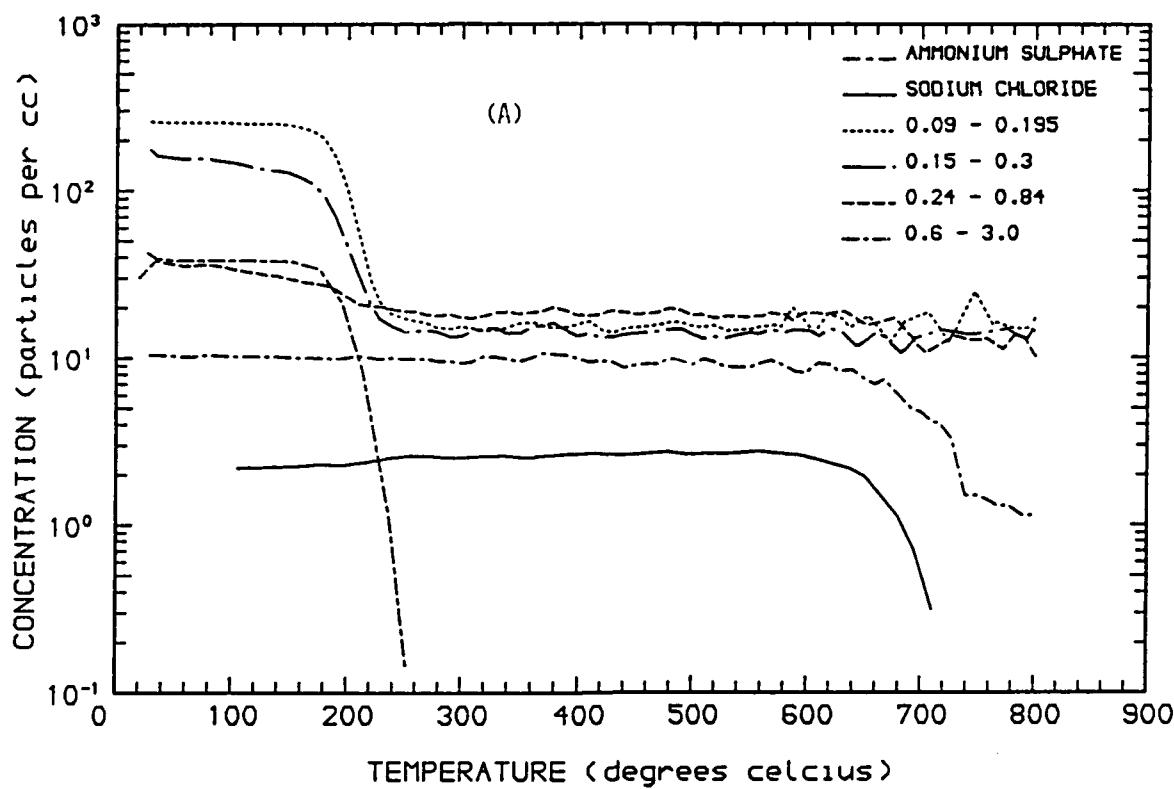


Fig. 5 (A) Temperature fractionation curves for field data at Mace Head site on June 2 1988, as compared to laboratory data for ammonium sulphate and sodium chloride over the temperature range from ambient to 800 °C.

(B) As per 5 (A) except for September 20 1988.

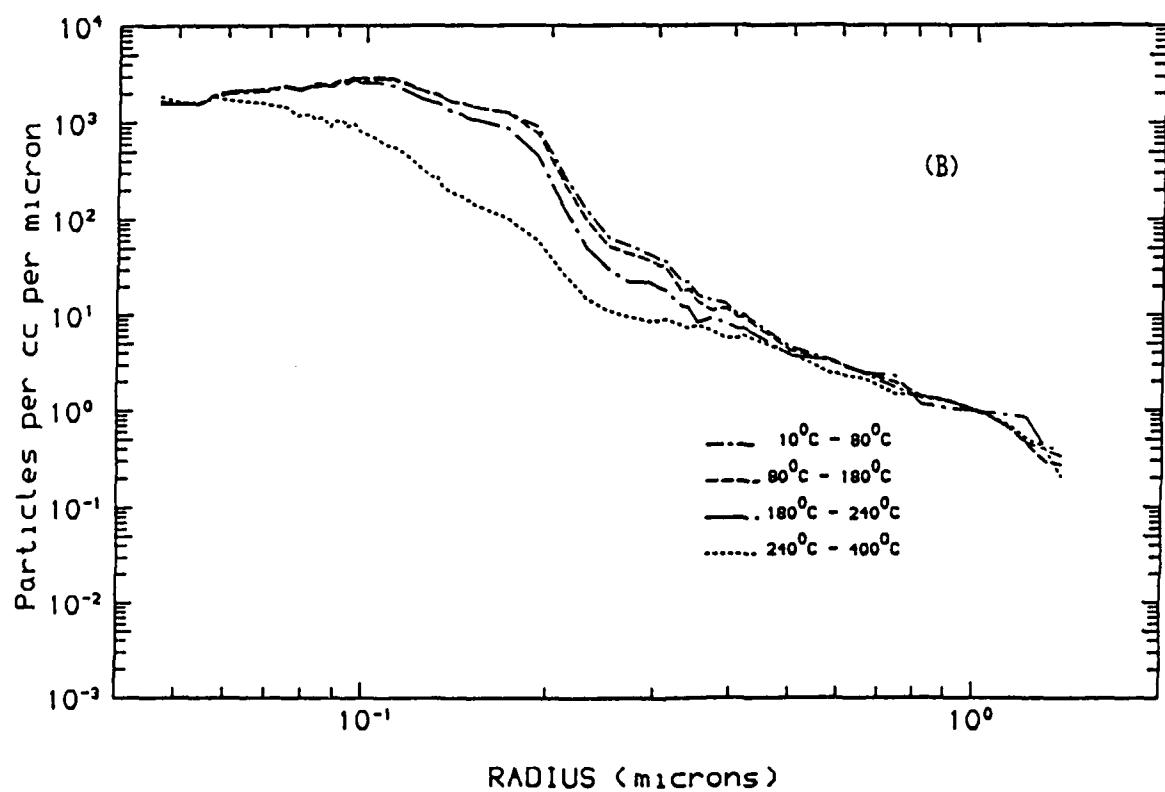
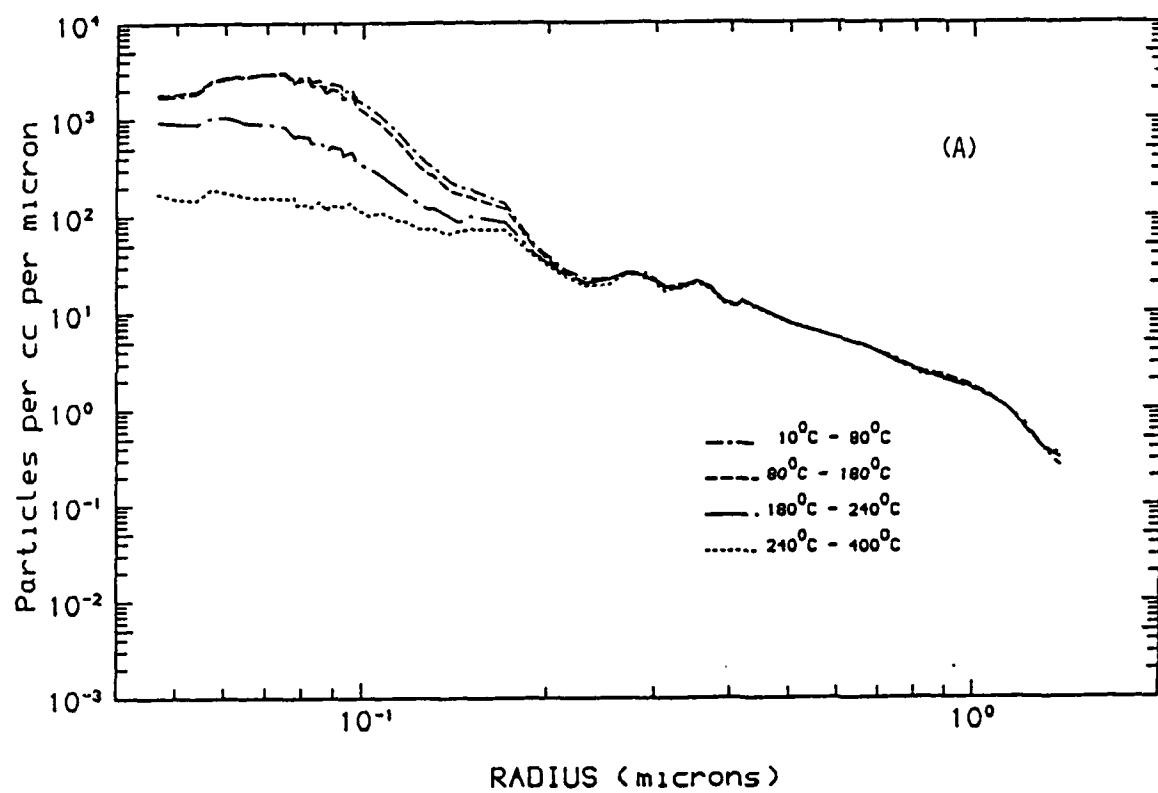


Fig. 6 Aerosol size distributions at Mace Head as a function of temperature up to 400 °C.

Measurement Periods: (A): June 1 1988; (B): Sept 20 1988.

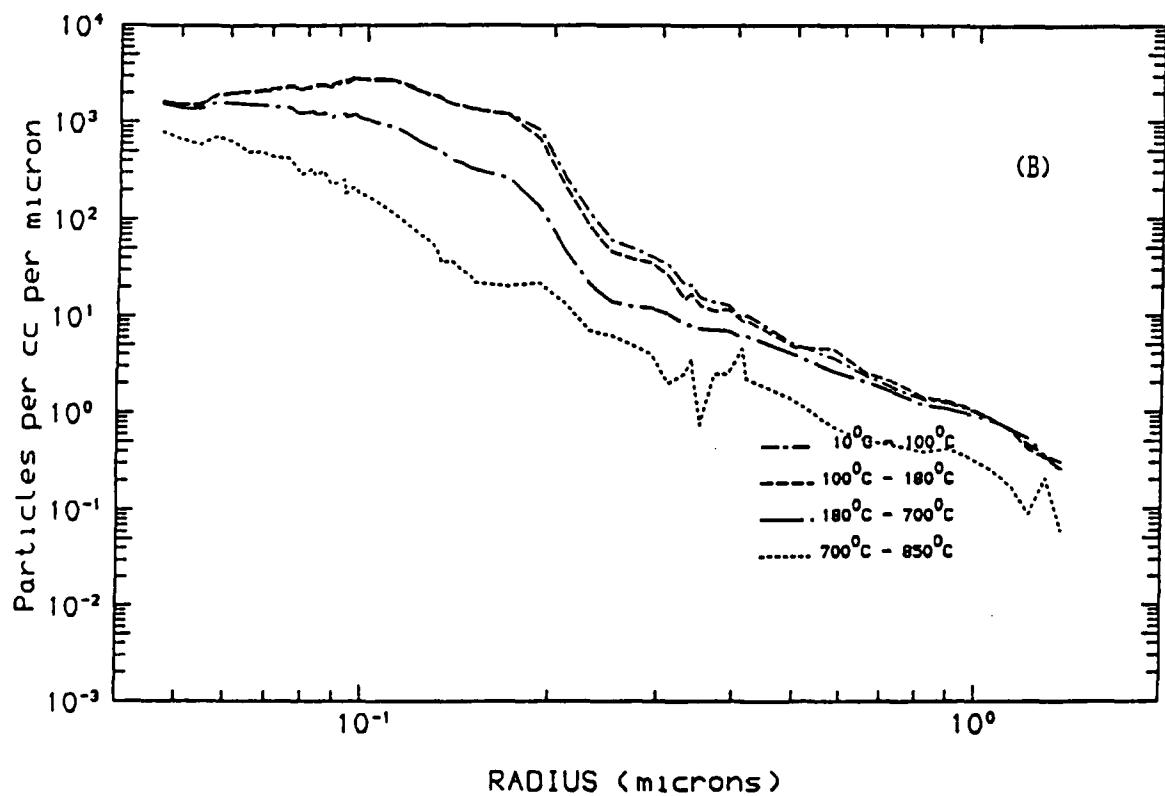
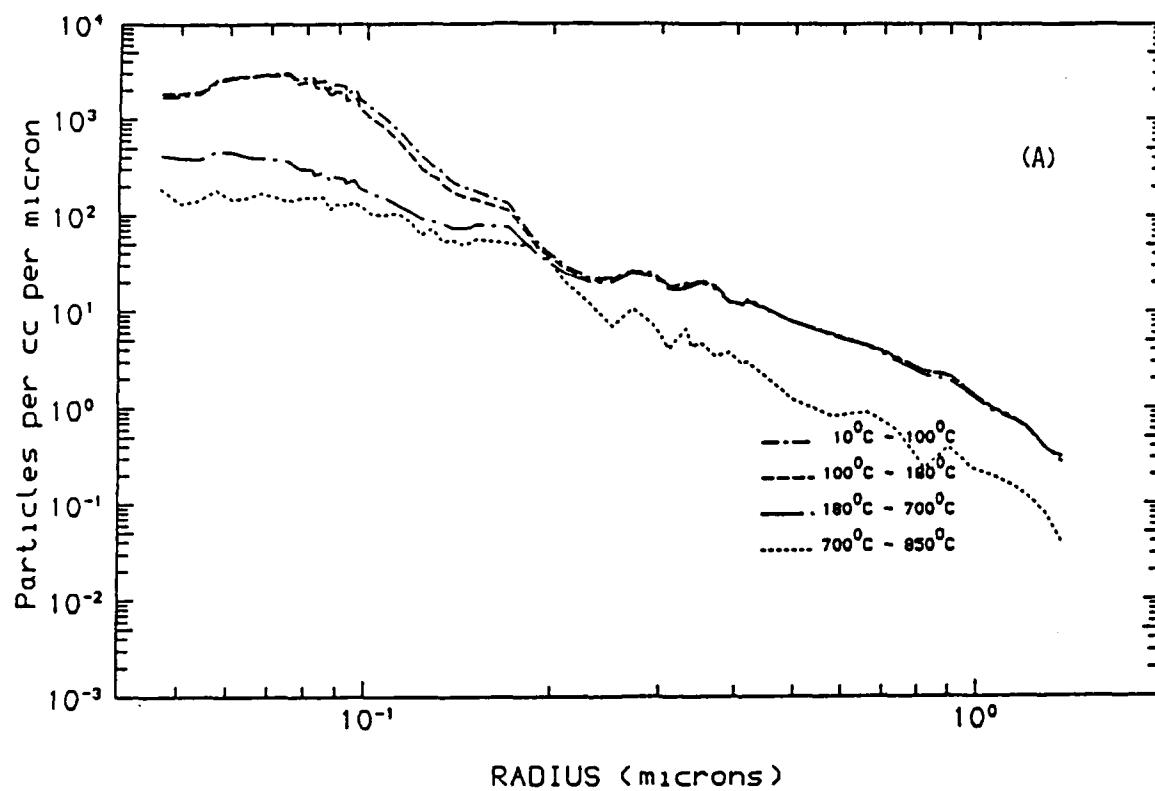


Fig. 7 Aerosol size distributions at Mace Head as a function of temperature up to 850 °C

Measurement periods: (A): June 1 1988; (B): Sept 20 1988

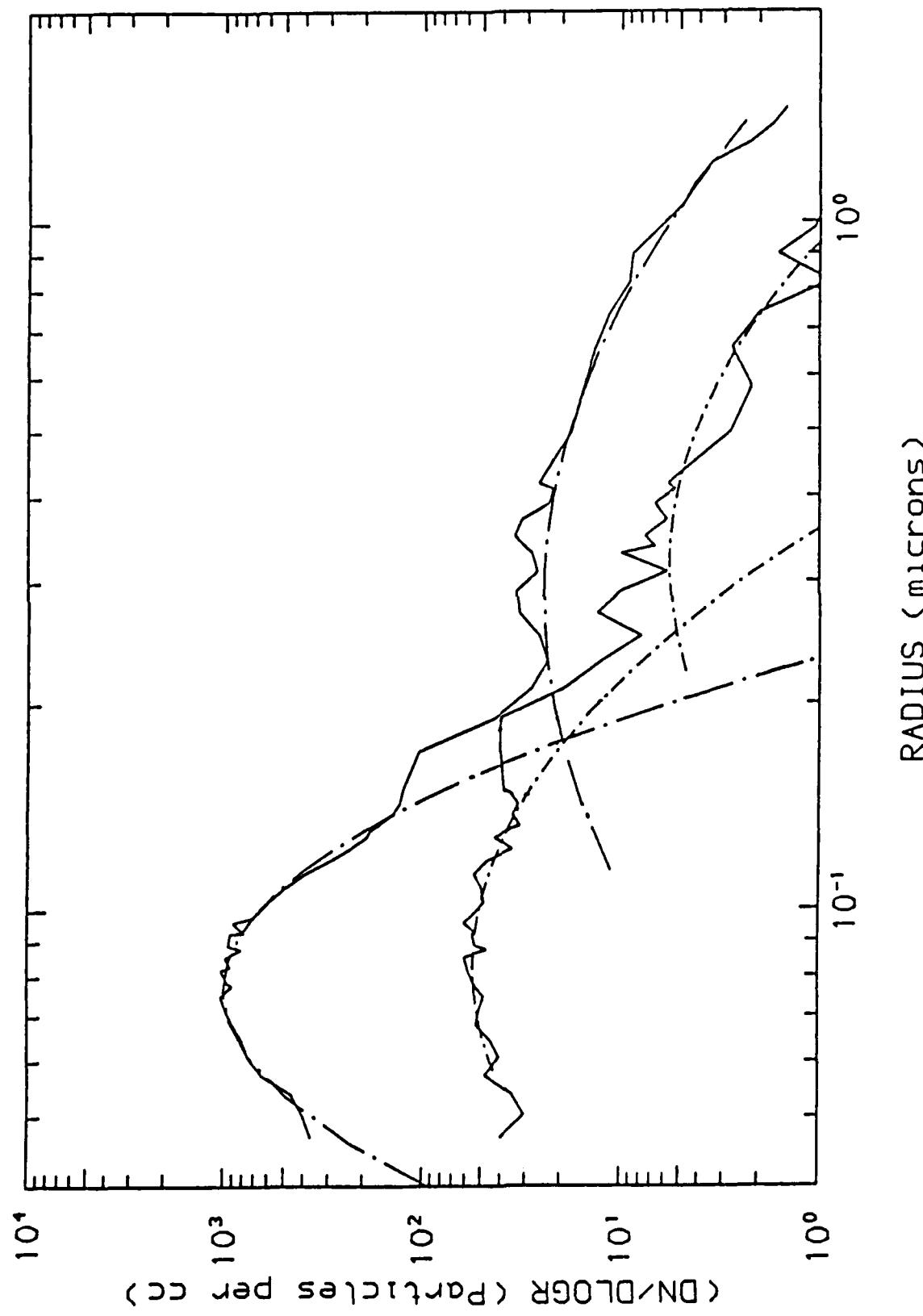


Fig. 8 Log normal size distribution fits to the ambient (upper curves) and residual (lower curves) aerosol for June 1-2, 1988. The log normal fitted parameters are given in Table 3.